

Annex 1
Amended claims (Clean version)

1. A method for the preparation of halogenated benzonitriles by vapour phase ammonoxidation of halogenated C₁ to C₆ alkyl benzenes, in the presence of water vapour, at a reaction temperature comprised in the range of 300 to 500 °C, using a three-component catalyst into a fixed bed reactor wherein said catalyst consists of a promoted VPO active phase provided on a carrier.

2. A method according to claim 1, wherein said reaction temperature is comprised in the range of 350 to 450 °C.

3. A method according to claim 1 or 2, wherein the residence time of the halogenated C₁ to C₆ alkyl benzene in said reactor is less than 10 seconds, preferably less than 8 seconds.

4. A method according to any of claims 1 to 3, wherein said halogenated C₁ to C₆ alkyl benzene is di- or tri-halogenated C₁ to C₆ alkyl benzene.

5. A method according to claim 4, wherein said di- or tri-halogenated C₁ to C₆ alkyl benzene is di-halogenated toluene.

6. A method according to claim 5, wherein said di-halogenated toluene is 2,6-dichlorotoluene.

7. A method according to any of claims 1 to 6, wherein said catalyst is provided on an Al₂O₃ carrier.

8. A method according to any of claims 1 to 6, wherein said catalyst is provided on a TiO₂ carrier.

9. A method according to claim 8, wherein said TiO₂ carrier consists of the anatase phase.

10. A method according to any of claims 1 to 9, wherein said catalyst comprises a V₁P_aM_bAl_cO_x or V₁P_aM_bTi_cO_x catalyst wherein M is chosen from the group comprising Cr, Fe, Co and Mo; a is 0.1 - 2.0; b is 0.002 - 1.0; c is 2.0 - 10.0, and x is determined by the valences of other component elements.

11. A method according to claim 10, wherein M is Co or Cr.

12. A method according to any of claims 1 to 11, whereby the catalyst is diluted with an inert medium in the ratio of 0.5 to 2.0 by weight with respect to the weight of said catalyst prior to its addition to the reactor.

13. A method according to claim 12 wherein said inert medium comprises corundum particles, porcelain beads, quartz beads, glass beads or the like.

14. A method according to any of claims 1 to 13, comprising the step of supplying a halogenated alkane to said reactor.

15. A supported and promoted VPO catalyst, suitable for use in a method according to any of claims 1 to 14, obtainable by a process comprising the steps of

- preparing a bulk VPO precursor ;
- impregnating said bulk VPO precursor with a promoter element in order to obtain a bulk promoted VPO precursor; and
- adding a carrier to said bulk promoted VPO precursor,
- and calcining the resultant mixture under a weakly oxidising atmosphere of O₂/N₂ to obtain a supported and promoted VPO catalyst,

whereby said catalyst is a V₁P_aM_bAl_cO_x or V₁P_aM_bTi_cO_x catalyst wherein M is chosen from the group comprising Cr, Fe, Co and Mo; a is 0.1 - 2.0; b is 0.002 - 1.0; c is 2.0 - 10.0, and x is determined by the valences of other component elements, characterized in that the catalyst contains mainly two phases under working conditions a (VO)₂P₂O₇ phase and a (NH₄)₂(VO)₃(P₂O₇)₂ phase.

16. A catalyst according to claim 15, wherein said carrier is an Al₂O₃ carrier.

17. A catalyst according to claim 15, wherein said carrier is a TiO₂ carrier.

18. A catalyst according to claim 17, wherein said TiO₂ carrier consists of the anatase phase.

19. A method for the preparation of a catalyst according to any of claims 15 to 18 comprising the steps of:
 - preparing a bulk VPO precursor;
 - impregnating said bulk VPO precursor with a promoter element in order to obtain a bulk promoted VPO precursor; and
 - adding a carrier to said bulk promoted VPO precursor,
 - and calcining the resultant mixture under a weakly oxidising atmosphere of O₂/N₂ to obtain a supported and promoted VPO catalyst.
20. A method according to claim 19 comprising the steps of:
 - refluxing of a vanadium source in the presence of alcohols to obtain a solution containing reduced vanadium species;
 - adding of a phosphorus source and refluxing said solution in order to obtain a bulk VPO precursor;
 - impregnation of said bulk VPO precursor with an alcoholic or aqueous solution of a promoter element to obtain a bulk promoted VPO precursor;
 - mixing said bulk promoted VPO precursor with γ -Al₂O₃ or TiO₂ carrier powder; and shaping and calcining the resultant mixture to obtain a supported and promoted VPO catalyst.
21. A method according to claim 19 or 20, wherein said vanadium source is V₂O₅ and said phosphorous source is o-H₃PO₄.
22. A method according to any of claims 19 to 21, wherein said promoter element comprises Cr, Fe, Co or Mo.
23. A method according to any of claims 19 to 22, wherein the ratio of said bulk promoted VPO precursor over said γ -Al₂O₃ or TiO₂ carrier powder is comprised in the range of 1:2 to 1:10, preferably 1:6.
24. A method according to any of claims 19 to 23, wherein the calcining step is performed at a temperature comprised in the range of 300 to 900°C for a period of 1 to 10 hours, under a calcining atmosphere.
25. A method according to any of claims 19 to 23, wherein the calcining step is performed at a temperature comprised in the range of 350 to 700°C for a period of 2 to 6 hours, under a calcining atmosphere of weak oxidising strength.
26. Use of a VPO catalyst according to any of claims 15 to 18 and prepared according to any of claims 20 to 26 in a vapour phase ammoxidation reaction.
27. Use of a VPO catalyst according to claim 26 in a vapour phase ammoxidation reaction according to any of claims 1 to 15.
28. Use of a VPO catalyst according to claim 27 for preparing 2,6-dichlorobenzonitrile from 2,6-dichlorotoluene in a vapour phase ammoxidation reaction.
29. Use of a VPO catalyst according to claim 28 in a vapour phase ammoxidation reaction of hetero aromatic hydrocarbons.